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Elemental Distribution of U and Ni in Contaminated Riparian Sediments

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Beamline(s): X26A

Introduction: Bulk characterization methods provide useful but indirect information on geochemical factors controlling the mobility and bioavailability of radionuclides and heavy metals in environmental media. For example, sequential extractions performed on contaminated riparian sediments from the Savannah River Site suggest strong associations between U and organic carbon and between Ni and Fe oxide minerals [1]; however, the limitations of such methods preclude identification of specific contaminant-mineral associations and heterogeneity at the soil mineral scale (i.e., µm's). Such detailed information is essential for developing a scientific basis for natural attenuation and other cost-effective environmental management and remediation strategies.

Methods and Materials: We used spatially resolved, synchrotron-based X-ray fluorescence (SXRF) mapping at NSLS microprobe beamline X26A to obtain spatial distributions and elemental associations of key contaminants in sediments (primarily U and Ni). Polished 30 µm thin-sections were prepared from intact, resin-embedded sediment cores sampled at locations exhibiting elevated U and Ni concentrations (up to 1000 mg kg^{-1}) and a range of organic carbon concentrations (~1 to 14% by wt.). A 17.2 keV beam was generated using a Si(111) monochromator and focused using Kirkpatrick-Baez mirrors onto the sample at 45º. Beam spot size varied between 10 x 15 and 15 x 30 µm. A Si(Li) detector was used for energy dispersive fluorescence measurements at 90º. Two-dimensional maps were obtained by rastering across the sample using a 10 to 15 µm step size.

Results: Spatially resolved SXRF mapping data generally corroborated previous results from indirect and bulk methods such as chemical extractions [1]. However, the ability to probe elemental compositions at the µm scale also demonstrated substantial heterogeneous partitioning of U and Ni in sediments. Similar results were reported previously for lower resolution mapping of intact soil particles with 50 to 300 µm beam sizes [2,3]. Frequently, U and Ni associations were mutually exclusive, i.e., U was associated with organic matter while Ni was associated with non-organic Fe-rich regions (Fig. 1). In other cases, U and Ni hotspots were spatially correlated. Observations of intact organic structures not enriched in anthropogenic metals, presumably root cross-sections, suggest a strong link between the degradation of organic matter and contaminant partitioning.

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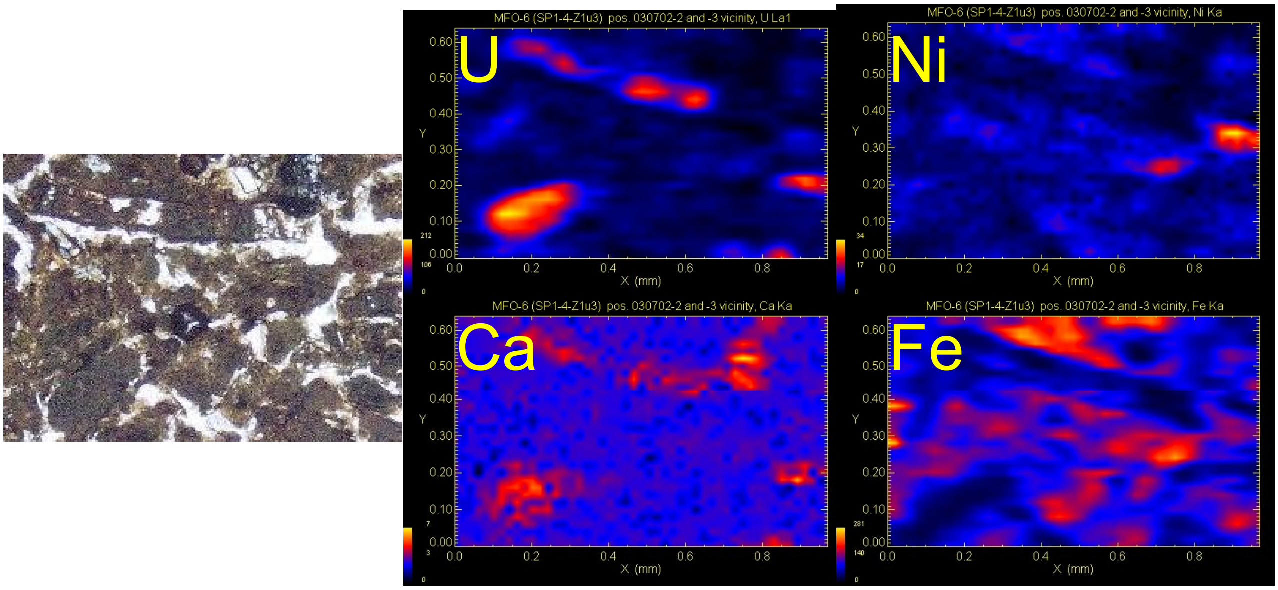

Figure 1. Photomicrograph (left) and 2D SXRF maps of U, Ni, Ca, and Fe (right) for a 30 μm sediment thin-section. U is localized primarily in Ca-rich organic features that are relatively low in Ni and Fe (e.g., linear feature in upper portion of field and elliptical feature in bottom left corner). Ni hotspots are generally associated with Fe-rich regions. Field corresponds to a 1 mm x 0.6 mm area. Step size for scan is 20 μm.